



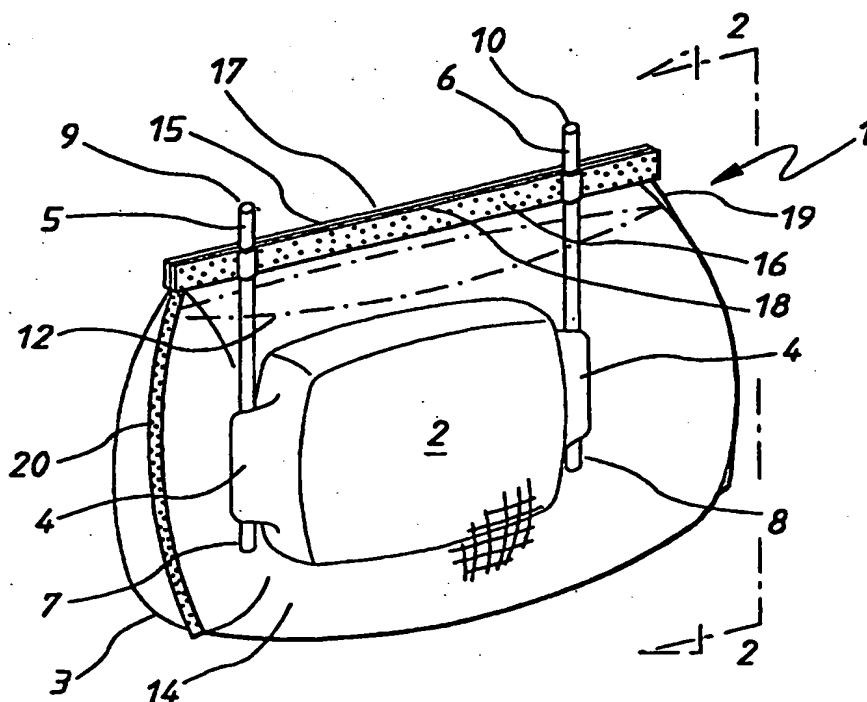
## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(21) International Application Number: PCT/AU99/00780 (22) International Filing Date: 16 September 1999 (16.09.99) (30) Priority Data: PP 5965 16 September 1998 (16.09.98) AU		(81) Designated States: AU, CA, JP, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  Published With international search report.
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(54) Title: A FLEXIBLE CHARGE STORAGE DEVICE

## (57) Abstract

A flexible charge storage device in the form of a supercapacitor (1) includes a flexible capacitive element (2) which is housed within a flexible plastics package (3). Element (2) includes a plurality of interleaved sheet electrodes having an insulator or separator therebetween and which are stacked in a face to face configuration. Alternate electrodes are interconnected by way of respective opposed tabs (4) which extend outwardly from element (2). Supercapacitor (1) also includes a first terminal (5) and a second terminal (6) which are respectively electrically connected with opposed tabs (4). More particularly, electrodes (5, 6) have first ends (7, 8) which are disposed within package (3), and respective second ends (9, 10) which are disposed outside of package (3). Ends (9, 10), in use, are connected to other components in the circuit of interest to allow electrical connection with the electrodes of element (2). Package (3) include a single folded laminar plastics sheet (14) which extends between two ends (15, 16), and which has edges (17, 18, 19 and 20). Ends (15, 16), as well as the portions of both sides (19, 20) which overlap, are fixedly and sealingly abutted against one another by way of heat welding or the like. Accordingly, electrolyte (12) is retained within package (3).



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**TITLE: A FLEXIBLE CHARGE STORAGE DEVICE****FIELD OF THE INVENTION**

The present invention relates to a charge storage device and in particular to a flexible charge storage device.

5       The invention has been developed primarily as a supercapacitor and will be described hereinafter with reference to that application. However, it will be appreciated that the invention is not limited to this particular field of use and is also suitable for other energy storage devices such as batteries, capacitors and the like.

**10   BACKGROUND OF THE INVENTION**

Known supercapacitors and other energy storage devices are provided in rigid containers or housings. These containers, particularly for supercapacitors offering large amounts of charge storage, are often fixedly mounted to other structures of the circuit or apparatus to which they are incorporated. The containers are generally bulky and  
15   necessitate prefabricated mounting points within the circuit or apparatus. Consequently, such circuits and apparatus do not easily accommodate a supercapacitor of differing dimension. In the event such a circuit requires a supercapacitor having different characteristics from that which was originally intended, the dimensions of the new capacitor must be very similar to the original.

20       It is an object of the present invention, at least in the preferred embodiments, to overcome or substantially ameliorate one or more of the disadvantages of the prior art, or at least provide a useful alternative.

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**DISCLOSURE OF THE INVENTION**

According to a first aspect of the invention there is provided a flexible charge storage device including:

a first sheet electrode having a first terminal extending therefrom;

5 a second sheet electrode disposed adjacent the first electrode and having a second terminal extending therefrom;

a porous separator disposed between the electrodes; and

a sealed package for containing the electrodes, the separator and an electrolyte, whereby the terminals extend from the package to allow electrical connection to the  
10 respective electrodes.

Preferably, each of the sheets includes two opposed sides, at least one of the sides of each sheet having a coating containing activated carbon.

Preferably also, the first electrode and the second electrode include respective first and second aluminium sheets. More preferably, the first and second sheets and the  
15 intermediate separator are together folded.

In a preferred form the charge storage device includes a plurality of first and second sheets and intermediate separators. More preferably, the sheets and intermediate separators are stacked. Alternatively, the sheets and the intermediate separators are wound together.

20 Preferably also, one of the length and breadth of the package is less than the respective length and breadth of the first sheet.

Preferably, the package includes a plurality of layers. More preferably, the layer of the package closest to the terminals is polyethylene. Even more preferably, the

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package is sealingly bonded to the terminals. Most preferably, the package is adhesively bonded to the terminals.

In a preferred form, the package includes polyethylene and the terminals are aluminium, wherein the adhesive bond is formed with an adhesive resin. In other

5 ~~embodiments use is made of an epoxy resin. In other preferred embodiments each~~  
terminal includes a respective plastics sleeve sealingly bonded thereto. In this  
embodiment the package is preferably sealingly engaged with the sleeves. In further  
embodiments a plastics layer is used in place of the sleeve.

Preferably, the sheets are abutted against the separator.

10 In a preferred form the device, when maintained at 80°C for 100 hours, retains at  
least 90% by weight of the electrolyte. Even more preferably, and under the same  
conditions, the device retains at least 95% by weight of the electrolyte.

According to a second aspect of the invention there is provided a method of  
producing a flexible charge storage device, the method including the steps of:

15 providing a first sheet electrode having a first terminal extending therefrom;  
disposing a second sheet electrode adjacent the first electrode, the second  
electrode having a second terminal extending therefrom;  
disposing a porous separator between the electrodes; and  
sealing the electrodes and the separator in a package containing an electrolyte,  
20 whereby the terminals extend from the package to allow electrical connection to the  
respective electrodes.

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Preferably, each of the sheets includes two opposed sides, and the method includes the further step of applying a coating containing activated carbon to at least one of the sides of each sheet.

Preferably also, the first electrode and the second electrode are respective first  
5 and second aluminium sheets. More preferably, the method includes the step of folding  
together the first and second sheets and the intermediate separator.

In a preferred form, the method includes the step of providing a plurality of first  
and second sheets and intermediate separators. More preferably, the method includes the  
step of stacking the sheets and intermediate separators. Alternatively, the method  
10 includes the step of winding together the sheets and the intermediate separators.

Preferably, the package includes a plurality of layers. More preferably, the layer  
of the package closest to the terminals includes polyethylene. In some embodiments that  
layer is coated with an ionomer and more preferably coated with SURLYN. Even more  
preferably, the method includes the step of sealingly bonding the package to the  
15 terminals. Most preferably, the method includes the step of adhesively bonding the  
package to the terminals.

Preferably also, the package includes polyethylene and the terminal is  
aluminium, wherein the method includes the step of forming the adhesive bond with a  
resin. Examples of commercially available and suitable resins are those which are  
20 marketed under the names NUCREL and PRIMACOR. In other embodiments use is  
made of an epoxy resin. In further embodiments the method includes the steps of  
sealingly bonding a respective plastics sleeve to each terminal and then sealingly  
engaging the package with the sleeves.

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In a preferred form the method includes the step of abutting the sheets against the separator.

## BRIEF DESCRIPTION OF THE DRAWINGS

5 A preferred embodiment of the invention will now be described, by way of example only, with reference to the accompanying drawings in which:

Figure 1 is a schematic perspective view of a charge storage device according to the invention;

Figure 2 is a cross section taken along line 2-2 of Figure 1; and

10 Figure 3 is a graph of the electrolyte weight loss for one of the supercapacitors according to the invention.

## PREFERRED EMBODIMENT OF THE INVENTION

Referring to Figures 1 and 2, a flexible charge storage device in the form a  
15 supercapacitor 1 includes a flexible capacitive element 2 which is housed within a flexible plastics package 3. Element 2 includes a plurality of interleaved sheet electrodes having an insulator or separator therebetween and which are stacked in a face to face configuration. Alternate electrodes are interconnected by way of respective opposed tabs 4 which extend outwardly from element 2.

20 Supercapacitor 1 also includes a first terminal 5 and a second terminal 6 which are respectively electrically connected with opposed tabs 4. More particularly, electrodes 5 and 6 have first ends 7 and 8 which are disposed within package 3, and respective second ends 9 and 10 which are disposed outside of package 3. Ends 9 and

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10, in use, are connected to other components in the circuit of interest to allow electrical connection with the electrodes of element 2.

In other embodiments the sheet electrodes are folded together, while in further embodiments they are wound together. In any event, element 2 is flexible in that it is

5 pliable. In the present embodiment element 2 includes four stacked aluminium sheets each coated on one side with a thin activated carbon layer. Each sheet is maintained in a spaced apart configuration from the adjacent sheet by way of intermediate porous separators. Each sheet electrode is about 40 mm x 40 mm and the total thickness of element 2 is about 0.5 mm.

10 In other embodiments both sides of the aluminium sheets include a carbon coating.

Terminals 5 and 6 are aluminium, although in other embodiments use is made different conductive materials.

Package 3 contains element 2 which is pre-soaked with an electrolyte 12.

15 Package 3 is impervious to the electrolyte and, as such, maintains it in contact with the element 2. In this case, the separator included within element 2 is porous to allow movement of ions between the adjacent electrodes. The electrolyte should be as conductive as possible and, in this embodiment, is an organic electrolyte based upon ethylene carbonate and dimethylcarbonate. In other embodiments other electrolytes are  
20 used such as tetraethylammonium tetrafluoroborate dissolved in propylene carbonate or acetonitrile at 0.5 to 1.5 M concentration. Other electrolytes would be known to those skilled in the art.



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Package 3 include a single folded laminar plastics sheet 14 which extends between two ends 15 and 16 and which has edges 17, 18, 19 and 20. Ends 15 and 16, as well as the portions of both sides 19 and 20 which overlap, are fixedly and sealingly abutted against one another by way of heat welding or the like. Accordingly, electrolyte

5 12 is retained within package 3. In the configuration shown the approximate length and breath dimensions of package 3 are both 50 mm.

Sheet 14, in this embodiment, includes four layers which have a combined thickness of about 100 microns. In other embodiments however, use is made of a sheet having a thickness in the range of 30 microns to 1 mm. Preferably, however, to optimise  
10 the flexibility and barrier properties of sheet 14 its total thickness is in the range of 50 microns to 200 microns.

In this embodiment the first or innermost layer of sheet 14 is polyethylene which is heat sealable and chemically unaffected by electrolyte 12. In other embodiments where use is made of alternative electrolytes it may be necessary to select an alternative  
15 innermost layer. The layer adjacent the polyethylene is ethylene vinyl alcohol which acts as a solvent and oxygen barrier. A layer of nylon is then utilised. This layer provides further solvent resistance and mechanical strength. The next or outermost layer in this embodiment is a barrier or protective layer and is preferably constructed from polyethylene. In other embodiments the outer layer is constructed of protective material  
20 other than polyethylene.

Not only does package 3 contain electrolyte 12, it prevents the ingress of water or other contaminants in to the electrolyte. Moreover, package 3 provides for a more effective sealing engagement with terminals 5 and 6. In this embodiment, where the

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terminals are made from aluminium and the inner layer of sheet 14 is polyethylene, the two are adhered together. Most preferably the adhesive is epoxy resin.

In another embodiment the packaging includes a surlyn coating and the resin selected from one of the commercially available resins such as the resins that are

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5 marketed under the names NUCREL and PRIMACOR.

To further enhance the sealing engagement some embodiments make use of pretreating the polyethylene surfaces by exposure to a corona discharge.

In alternative embodiments respective plastics sleeves or layers are initially bonded to terminals 5 and 6. The preferred sleeve is made from polyethylene, although  
10 other embodiments utilise other polymers such as polyethylene acrylic acid. The sleeves are heat bonded to the terminals. Thereafter, package 3 is heat sealed to the sleeves to form a sealed container for electrolyte 12.

In some embodiments the sleeve or layer is adhesively bonded to the terminals with a resin.

15 A number of embodiments of the invention were tested for electrolyte retention at elevated temperatures. The first test was conducted using a package of the preferred embodiment measuring 50 mm x 50 mm and containing dimethylcarbonate solvent. The supercapacitor was maintained at 80° C and the weight loss of electrolyte was found to be 1.9%, 3.2%, 7.3% and 13.1% at 100 hours, 200 hours, 600 hours and 1000 hours  
20 respectively.

The second test was conducted with another supercapacitor that provides about 15 Farads at 2.5 Volts. The electrolyte weight loss for the capacitor is plotted against time and illustrated in Figure 3. This supercapacitor has the following structure:

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four aluminium sheets, each being about 70 mm by 60 mm and including respective opposed carbon coatings;

two terminals being integrally formed with and extending away from the respective sheets;

5 ~~a separator for maintaining the sheets being in a fixed spaced apart configuration;~~

a multi-layer laminate packaging including an aluminium layer and an outer surlyn layer;

an acetonitrile solvent containing a 1 M solution of tetraethylammonium tetrafluoroborate;

10 a PRIMACOR resin for sealing the package against itself and the terminals;

a total weight of about 6 grams; and

overall dimensions of about 80 mm x 70 mm x 2mm.

In some embodiments package 3 includes different or additional layers.

Different polymers are used if specific operating temperatures are envisaged.

15 Supercapacitor 1 weighs approximately 5 grams and includes a capacitance of about 3 Farads at 2.5 Volts. As would be appreciated by a skilled addressee many other configurations are available. The present configuration, however, is particularly suited to mobile communications, self propelled toys, and automotive applications. In one specific embodiment, supercapacitor 1 is placed in parallel with a standard mobile

20 telephone battery. Such an arrangement not only extends the life of the battery but will quickly recharge. The compact and flexible nature of the capacitor and its package 3 allows them to be placed in confined spaces and in many different configurations.

Moreover, so far as the space available permits, additional like capacitors can be added

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in parallel to further the available charge storage capacity for the electronic device. In some cases, capacitor 1 is contained within the housing of the mobile telephone, while in other embodiments it is contained within the housing of the battery. However, in still further embodiments, supercapacitor 1 is maintained in the cavity between the mobile

5 telephone and the battery and, as such can be retro fitted to existing mobile telephones.

In the event the mobile telephone includes the necessary voltage regulation circuitry it can be powered by the supercapacitor only.

Another application of supercapacitor 1 is in combination with the laptop computers and other electronic equipment. Other applications include portable power  
10 tools and other cordless electric devices. Again, although supercapacitor 1 will advantageously operate in parallel with an existing battery, it is, in some embodiments, utilised as the sole power source.

Although the invention has been described with reference to specific examples, it will be appreciated by those skilled in the art that it may be embodied in many other  
15 forms.

**CLAIMS**

1. A flexible charge storage device including:
  - a first sheet electrode having a first terminal extending therefrom;
  - a second sheet electrode disposed adjacent the first electrode and having a second
  - 5 terminal extending therefrom;
  - a porous separator disposed between the electrodes; and
  - a sealed package for containing the electrodes, the separator and an electrolyte,
  - whereby the terminals extend from the package to allow electrical connection to the
  - respective electrodes.
- 10 2. A device according to claim 1 wherein each of the sheets includes two opposed sides, at least one of the sides of each sheet having a coating containing activated carbon.
3. A device according to claim 1 or claim 2 wherein the first electrode and the second electrode include respective first and second aluminium sheets.
4. A device according to claim 3 wherein the first and second sheets and the
- 15 intermediate separator are together folded.
5. A device according to claim 3 including a plurality of like first sheets, a plurality of like second sheets and a plurality of intermediate separators.
6. A device according to claim 3 or claim 5 wherein the sheets and intermediate separators are stacked.
- 20 7. A device according to claim 3 or claim 5 wherein the sheets and the intermediate separators are wound together.

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8. A device according to any one of the preceding claims wherein one of the length and breadth of the package is less than the respective length and breadth of the first sheet.

9. A device according to any one of the preceding claims wherein the package

5 includes a plurality of layers.

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10. A device according to claim 9 wherein the layer of the package closest to the terminals includes polyethylene.

11. A device according to claim 10 wherein the layer closest to the terminals includes an ionomer coating.

10 12. A device according to claim 11 wherein the ionomer coating is a SURLYN coating.

13. A device according to claim 9 wherein the package is sealingly bonded to the terminals.

14. A device according to claim 11 wherein the package is adhesively bonded to the  
15 terminals.

15. A device according to claim 13 wherein the package includes a ionomer coating and the terminals are aluminium, wherein the adhesive bond is formed with an adhesive resin.

16. A device according to claim 13 wherein the ionomer coating is a SURLYN  
20 coating.

17. A device according to claim 15 wherein the resin is an epoxy resin.

18. A device according to claim 15 wherein each terminal includes a respective plastics sleeve sealingly bonded thereto.

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19. A device according to claim 18 wherein the package is sealingly engaged with the sleeves.

20. A device according to claim 15 including a plastics sheet disposed in sealing abutment with the terminals and the package.

5 21. A device according to any one of the preceding claims wherein the sheets are abutted against the separator.

22. A device according to any one of the preceding claims which, when maintained at 80°C for 100 hours, retains at least 90% by weight of the electrolyte.

23. A device according to claim 22 which, under the same conditions, retains at least  
10 95% by weight of the electrolyte.

24. A device according to any one of claims 1 to 21 which, when maintained at 70°C for 1000 hours, retains at least 99% by weight of the electrolyte.

25. A method of producing a flexible charge storage device, the method including the steps of:

15 providing a first sheet electrode having a first terminal extending therefrom;  
disposing a second sheet electrode adjacent the first electrode, the second electrode having a second terminal extending therefrom;  
disposing a porous separator between the electrodes; and  
sealing the electrodes and the separator in a package containing an electrolyte,  
20 whereby the terminals extend from the package to allow electrical connection to the respective electrodes.

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26. A method according to claim 25 wherein each of the sheets includes two opposed sides, and the method includes the further step of applying a coating containing activated carbon to at least one of the sides of each sheet.

27. A method according to claim 25 or claim 26 wherein the first electrode and the

~~5 second electrode are respective first and second aluminium sheets and the method~~

includes the step of folding together the first and second sheets and the intermediate separator.

28. A method according to claim 27 including the step of providing a plurality of like first sheets, a plurality of like second sheets and a plurality of intermediate separators.

10 29. A method according to claim 28 including the step of stacking the sheets and intermediate separators.

30. A method according to claim 28 including the step of winding together the sheets and the intermediate separators.

31. A method according to any one of claims 25 to 28 wherein the package includes  
15 a plurality of layers one of which is polyethylene and the method includes the step of disposing the polyethylene layer of the package closest to the terminals.

32. A method according to claim 31 including the step of sealingly bonding the package to the terminals.

33. A method according to claim 32 including the step of adhesively bonding the  
20 package to the terminals.



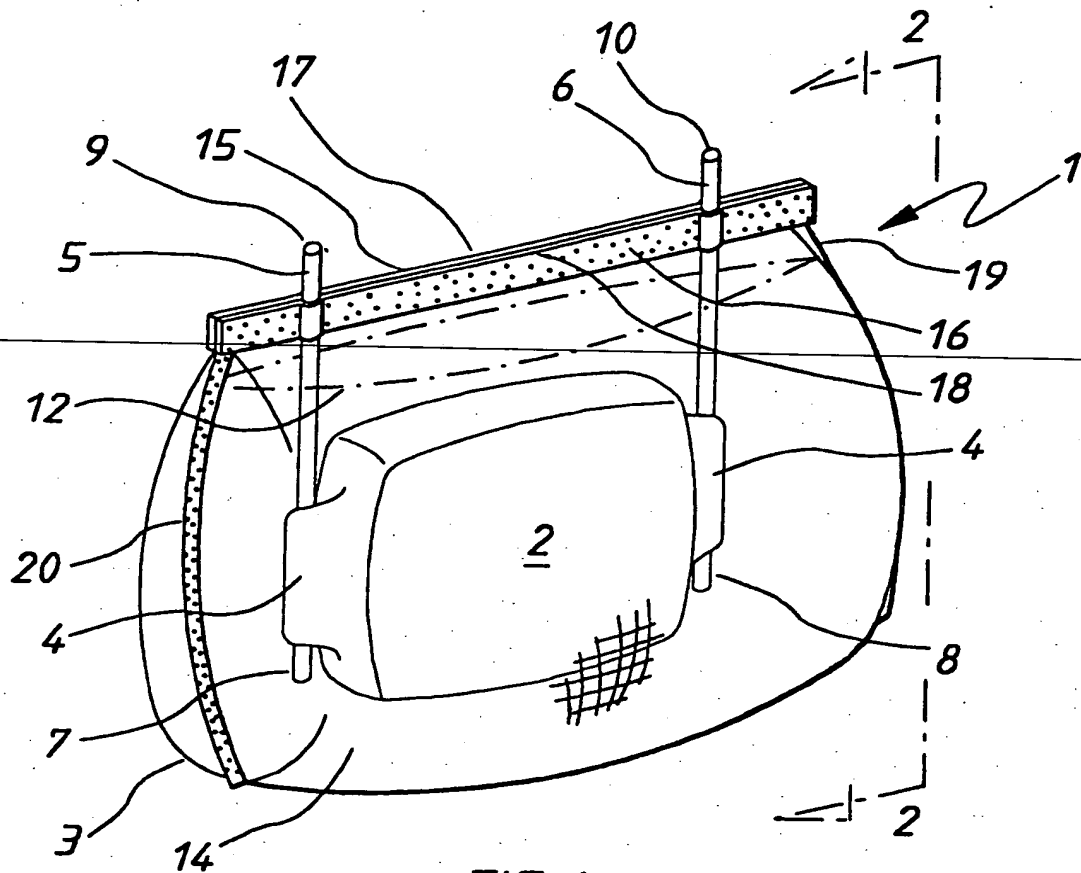


FIG. 1

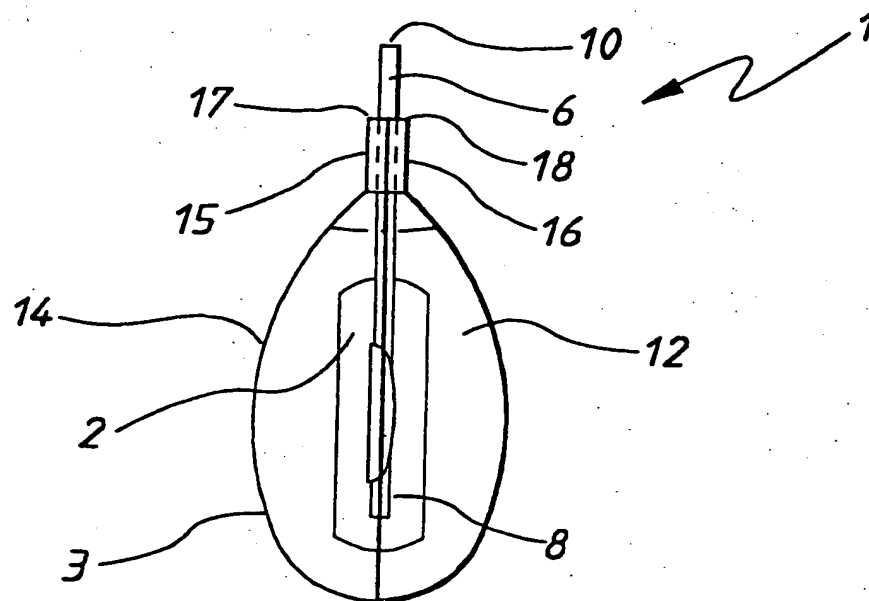


FIG. 2

electrolyte weight loss at 70°C

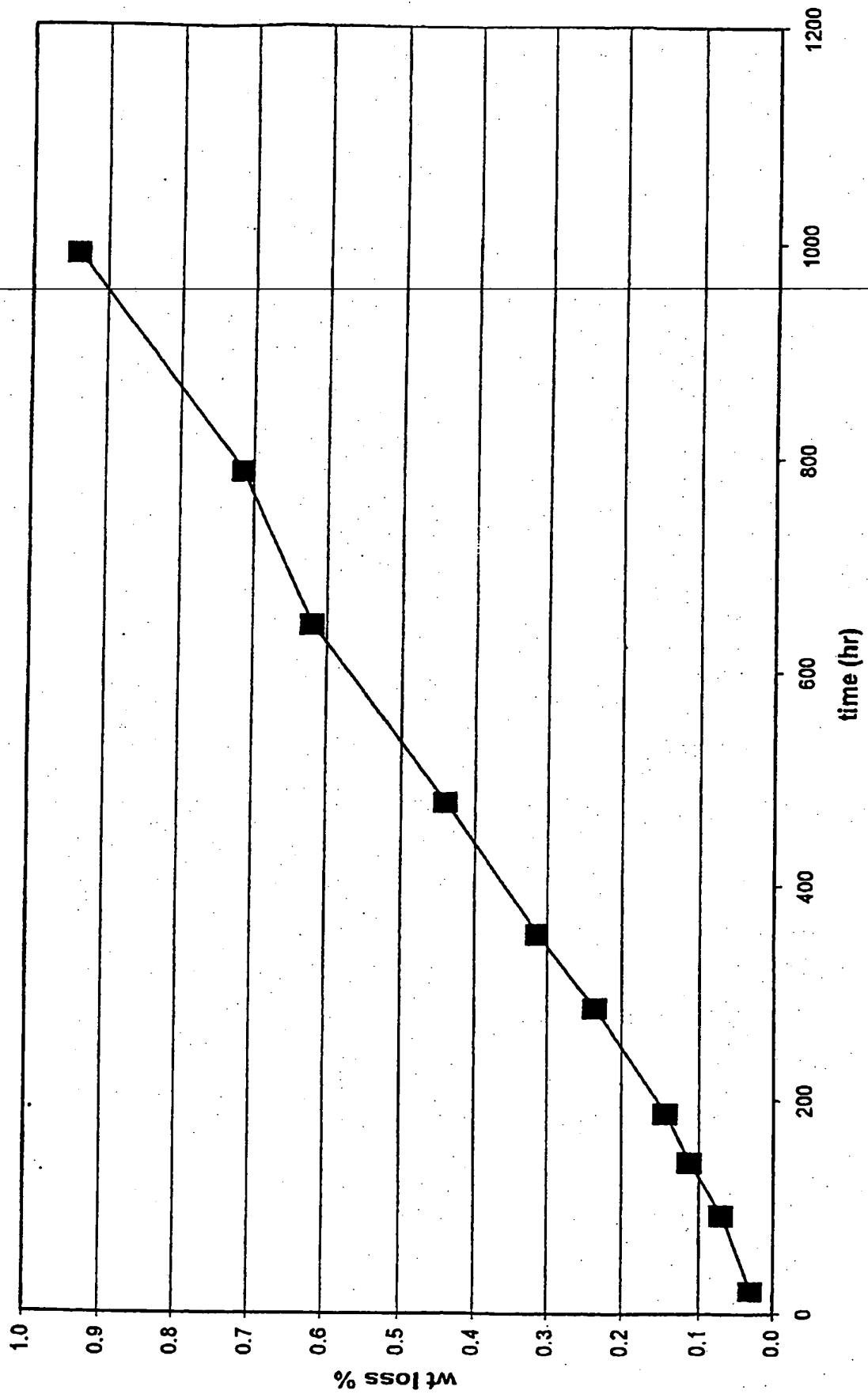


Figure 3

## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/AU 99/00780

**A. CLASSIFICATION OF SUBJECT MATTER**

Int Cl<sup>6</sup>: H01G 4/26, 9/058

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC H01G 9/-, 4/26, 13/-

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
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Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
WPAT and JPAT: (capacitor# or condensor#) and electrolyt: and (flex: or fold: or bend: or bent or deform:) and (sheet or plan: or flat)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	Derwent Abstract Accession No. 33323 D/19, Class A85 L03, JP 56-027914 A (MATSUSHITA ELECTRIC) 18 March 1981 See fig 4.	1-4,7,9,25
E,X	WO 99/53510 (ENERGY STORAGE SYSTEMS LTD et al) 21 October 1999	1,25
X	Derwent Abstract Accession No. 92-246734/30, Class V01, JP 4-167415 A (ASAHI GLASS) 15 June 1992	1

☒ Further documents are listed in the  
continuation of Box C

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International application No.

PCT/AU 99/00780

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	Derwent Abstract Accession No. 92-246735/30, Class V01, JP 4-167416 A (ASAHI GLASS) 15 June 1992	1
X	GB 2046019 A (SPRAGUE ELECTRIC) 5 November 1980 See figures and abstract.	1
X	GB 2046020 A (SPRAGUE ELECTRIC) 5 November 1980 See figures and abstract.	1
A	Derwent Abstract Accession No. 95-64485/09, Class V01, JP 6-342739 A (MATSUSHITA DENKI SANGYO) 13 December 1994	

# INTERNATIONAL SEARCH REPORT

## Information on patent family members

International application No.  
PCT/AU 99/00780

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report			Patent Family Member					
JP	56-027914	NONE						
JP	4-167415	NONE						
JP	4-167416	NONE						
GB	2046019	CA	1136232	US	4267566	JP	55-133531	
GB	2046020	US	4267565	CA	1118853	JP	55-132031	
JP	6-342739	NONE						

END OF ANNEX

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